Defense Technical Information Center Compilation Part Notice

ADP012289

TITLE: Magnetic and Microstructural Aspects of the Bulk Metallic Glassy Materials Nd60Fe30Al10

DISTRIBUTION: Approved for public release, distribution unlimited

This paper is part of the following report:

TITLE: Applications of Ferromagnetic and Optical Materials, Storage and Magnetoelectronics: Symposia Held in San Francisco, California, U.S.A. on April 16-20, 2001

To order the complete compilation report, use: ADA402512

The component part is provided here to allow users access to individually authored sections of proceedings, annals, symposia, etc. However, the component should be considered within the context of the overall compilation report and not as a stand-alone technical report.

The following component part numbers comprise the compilation report:
ADP012260 thru ADP012329

Mat. Res. Soc. Symp. Proc. Vol. 674 © 2001 Materials Research Society

Magnetic and Microstructural Aspects of the Bulk Metallic Glassy Materials $Nd_{60}Fe_{30}Al_{10}$

N. H. Dan, N. X. Phuc, V. H. Ky, N. M. Hong
Institute of Materials Science, NCST, Hoang Quoc Viet Str., Hanoi, Vietnam

N. Chau, N. H. Luong, C. X. Huu
Center for Materials Science, Department of Physics, Hanoi National University, Nguyen Trai Str., Hanoi, Vietnam.

R. W. McCallum, M. J. Kramer, A. S. O'Connor, K. W. Dennis
Ames Laboratory, USDOE and Department of Materials Science and Engineering,
Iowa State University, Ames, IA 50011, USA

L. H. Lewis
Materials and Chemical Sciences Division, Energy Sciences and Technology Dept., Brookhaven National Laboratory, Upton, New York 11973-5000, USA

L. D. Tung
International Training Institute for Materials Science, 1 Dai Co Viet Str., Hanoi, Vietnam

ABSTRACT

The ferromagnetic bulk metallic glass (BMG) $Nd_{60}Fe_{30}Al_{10}$ system exhibits extremely large coercivities at low temperature and moderate coercivities near room temperature. The magnetic hardness, as best evidenced by the onset of magnetic irreversibility, was studied in bulk suction-cast and melt-spun alloys with the nominal composition $Nd_{60}Fe_{30}Al_{10}$. Systematic x-ray diffraction studies of the degree of crystallinity performed as a function of position within the bulk suction-cast samples is found to correlate with the variation in the room-temperature magnetic hysteresis character. X-ray diffraction data clearly shows the presence of both crystallites and amorphous material on the samples' outmost surfaces; the amorphous phase content increases with distance into the cast sample. These results underscore the importance of solidification conditions and attendant nanophase selection, on the resultant magnetic properties of this class of alloys.

INTRODUCTION

The ferromagnetic bulk metallic glass composition $RE_{60}Fe_{30}Al_{10}$ (RE=Nd or Pr) has generated considerable interest of both applied and fundamental nature by virtue of its appreciable coercivity at room temperature [1]. The presence and magnitude of this reported coercivity, up to 0.4 T at room temperature, is an apparent contradiction to the conventional understanding of the relationship between nanostructure and coercivity in nominally amorphous materials. An additional challenge associated with study of this class of materials is the difficulty of reproducing the results reported in various laboratories around the world [2-6]. This latter challenge indicates that the material preparation conditions, such as the purity of the starting materials, the prealloy state [7] and the solidification condition, all influence the resultant magnetic properties. To further clarify the relationship between microstructure and magnetic properties in this class of materials, systematic studies of the position-dependent degree of crystallinity with the suction-cast rods were correlated with the room-temperature magnetic response. The considerable variation in magnetic response with degree of crystallinity underscores the importance of solidification conditions, and attendant nanophase selection, on the resultant magnetic properties of this class of alloys.

EXPERIMENTAL DETAILS

Investigations into both the temperature- and field-dependent character of the coercivity and into the relationships between the microstructure and the room-temperature coercivity were carried out on bulk suction-cast and melt-spun forms of $Nd_{60}Fe_{30}Al_{10}$ made from the elements Nd (2-9's purity), Fe (4-9's purity) and Al (4-9's purity). The alloy ingots were used to prepare two forms of the samples: melt-spun ribbons were formed on a copper single-wheel roller operated at various circumferential speed and hence with with various quenching rates, and rods that were suction-cast into copper molds of various rectangular cross-sections. The dimensions of the rod samples were height $z_0 \approx 60$ mm, width $y_0 \approx 10$ mm and thicknesses $x_0 \approx 1$, 1.5 or 3 mm. All rods were sectioned and polished parallel to the (y_0,z_0) sample plane. The phase constitution and degree of crystallinity of both forms of sample were assessed with room-temperature Cu-K_α x-ray diffraction (XRD). While materials melt-spun at high circumferential wheel speeds proved to be x-ray amorphous, the most meaningful data was obtained from partially-crystalline samples quenched at lower wheel speeds. The results obtained from the partially-crystalline samples were extrapolated to explain the results manifest in more amorphous samples.

The magnetic characterization was performed in an Ar atmosphere with a vibrating sample magnetometer both on the melt-spun ribbons and on 1 mm^3 cubic-shaped specimens taken from the center of the thickest cast rod. The applied field was in the range -15 kG $\leq H_{appl} \leq +15$ kG and was applied parallel to the long axis of the ribbons; the temperature range of measurement varied from 100 K – 600 K. The magnetic response of the bulk cube specimen showed no dependence on orientation with respect to the magnetic field. After raising the temperature well above the temperature range where the materials exhibit hysteresis, magnetization versus temperature measurements were made using both zero-field-cooled (ZFC) and field-cooled (FC) measurements which diverge at a temperature T_{irr} marking the appearance of irreversible magnetic behavior, Fig. 1. The divergence of the ZFC and FC measurements at T_{irr} is most easily understood in terms of the normalized field, $H_n=H_{appl}/H_c(T)$ where H_{appl} is the applied field and $H_c(T)$ is the temperate-dependent coercivity of the magnetically hardest phase present in the sample. In the ZFC measurement, the field is applied at the lowest temperature and the sample magnetization follows the initial magnetization curve. As the temperature is increased at fixed applied filed H_{app}, the normalized field H_n increases as $H_c(T)$ decreases so that the magnetization response moves to a higher value along the temperature-dependent initial magnetization curve. For the FC measurement, H_n starts at a value which effectively saturates the sample and then H_n decreases as $H_c(T)$ increases with decreasing temperature so that the magnetization moves down the first-quadrant demagnetization curve. Thus the difference between the ZFC and FC curves represents the difference between the initial magnetization curve and the demagnetization curve for the applied field and temperature. In other words T_{irr}, the temperature at which $M_{ZFC} = M_{FC}$, corresponds to the temperature where the width of the hysteresis loop closes at H_{appl}. For a hypothetical sample consisting of a paramagnetic phase combined with a hard magnetic phase with coercivity $H_{c(Hard)}$ that is independent of temperature, H_{irr} is also independent of temperature while it is clear that H_c of the sample is strongly dependent on temperature do to the temperature dependence of the paramagnetic fraction. Thus, due to the multiple magnetic phase behavior of the samples studied, T_{irr} is considered to be a more consistent measure of magnetic hysteresis than the value of the coercivity. T_{irr} was followed as a function of applied field for a melt-spun ribbon sample quenched at 30 m/sec and for a sample of the drop-cast material of 1 mm thickness, Fig. 1 inset.

RESULTS AND DISCUSSION:

The ZFC and FC magnetization measurements were performed on both melt-spun and bulk cast rod specimens of $Nd_{60}Fe_{30}Al_{10}$; representative results obtained at an applied field of 0.5 kG are displayed in Fig. 1. Strictly speaking the measurement fields are not equivalent due to the large differnce in demagnetization factor between the two samples. However this effect should lower T_{irr} in the bulk sample so that the actual difference in curves is enhanced over that which is measured. The departure from magnetic reversibility occurs at approximately 450 K for the bulk cast sample and around 325 K for the melt-spun ribbon at the measuring field of 0.5 kG. The difference in the onset of the magnetic irreversibility temperature in the two samples must be due to microstructural differences (including significant fluctuations in chemical composition in the disordered matrix) originating from the two solidification methods, as the nominal compositions

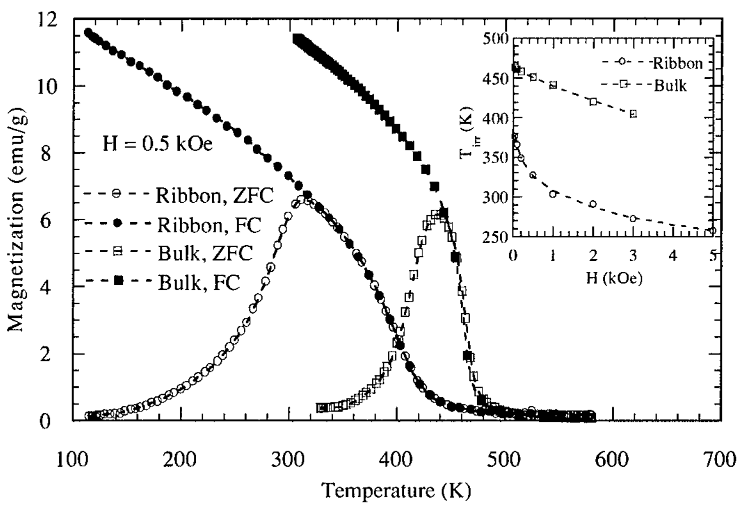

Figure 1. Magnetization data obtained at 0.5 kG in the zero-field-cooled (M_{ZFC}) and the field-cooled (M_{FC}) states from samples of the $Nd_{60}Fe_{30}Al_{10}$ in the form of ribbons melt-spun at 30 m/sec and cast rods of 1 mm diameter. The inset shows the field dependence of the onset temperature (T_{irr}) of the magnetic irreversibility temperature.

are the same. The origins of the differences in the onset of magnetic irrevsibility noted in the two different forms of the composition $Nd_{60}Fe_{30}Al_{10}$, Fig. 1, were pursued with systematic studies of the variation of the microstructure and magnetic properties in the bulk rods. Representative x-ray diffraction results are presented in Figs. 2 and 3. Significant varia- tion the of microstructure with position within the sample appears in ribbon melt-spun at 25 m/s with thickness ~ 40 μm. The surfaces of the ribbons were investigated separately: the "wheel side" surface, which describes the ribbon surface that was in direct contact with the quenching wheel and the "free side" ribbon surface that was exposed to the quenching atmosphere. As shown in Fig 2, the broad diffraction peak observed for the wheel-side surface at $2\theta \approx 30^{\circ}$ is sharper than the corresponding peak for the ribbon free-side surface. This observation motivated study of the position-dependent microstructure and corresponding magnetic properties for samples of millimeter-scale thickness.

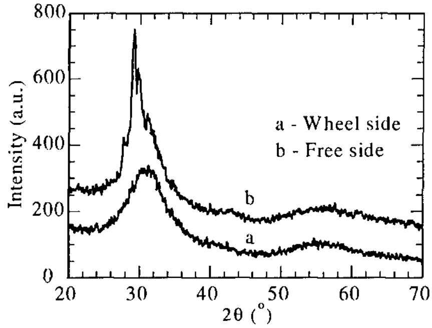

Figure 2. XRD patterns of Nd-Fe-Al ribbon melt-spun at 25 m/s.

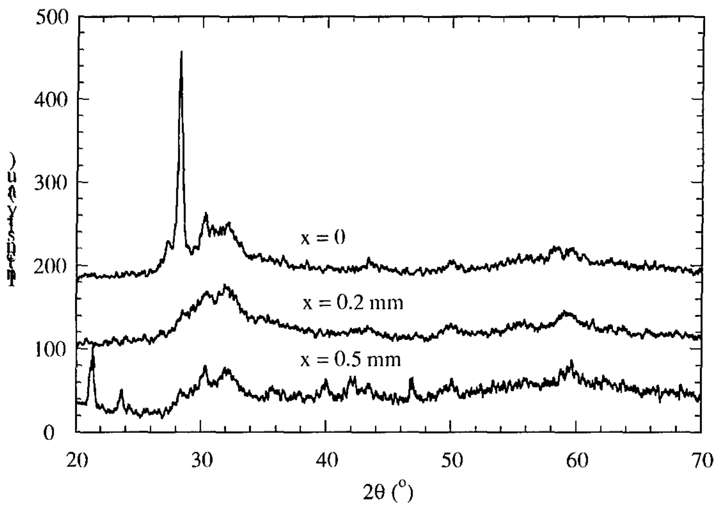

Figure 3. XRD patterns taken from cast rod of diameter 1 mm at depths x = 0 (cast surface), x = 0.2 mm and x = 0.5 mm. Note that the sample assumes a more amorphous character with increasing depth in the rod.

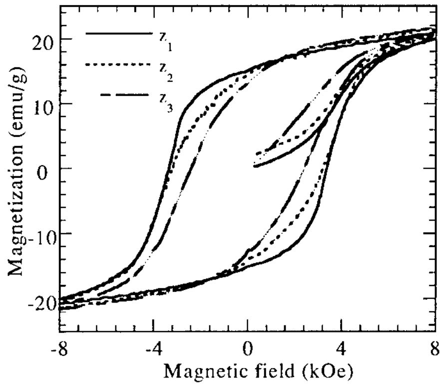

Figure 4a). Room-temperature hysteresis loops of 1-mm thick cast rod sample for specimens sectioned at various height z positions.

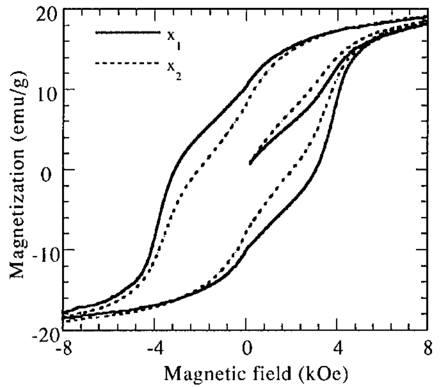

Figure 4b). Room-temperature hysteresis loops for specimens sectioned from the outermost (x_1) and innermost (x_2) position of 3-mm-thick cast rod.

XRD scans reveal a variation of crystallinity with respect to position within the suction-cast samples, Figure 3. The original cast surfaces of all rods are revealed to be partially crystalline. Systematic magnetic investigation from samples taken from at a variety of depths within the cast samples show a variation of hysteretic character that reflects the variation of crystallinity, as discussed above. The magnetic data of Fig. 4a) contradict the XRD data of Fig. 3 by demonstrating the existence of small but signficant differences in the microstructure along the length (z-direction) of the cast rod. More pronounced differences in the magnetic properties are found along the sample thickness (Fig. 4b)) as expected from the XRD data. An interesting question is why the outer sections of the cast specimen exhibit a larger coercivity than do inner sections (Fig. 4b)). Clearly the data presented above concerning different forms (*i.e.*, melt-spun ribbons *vs.* cast rods) of the BMG composition $Nd_{60}Fe_{30}Al_{10}$ suggest that the solidification conditions, and attendant nanophase selection, greatly influence the magnetic properties. Heat flow patterns are quite different in the various casting techniques investigated during the course of this study. The presence of significant crystallization on the free surface of the single-roller melt-spun ribbons (Fig.2) indicates that the cooling rate at this surface is suffciently slow to allow nucleation and growth of the pre-existing nanoscale clusters [7]. Analogously, the initial cooling rate near the mold wall is insufficient to prevent nucleation and growth at the starting composition. In the bulk samples there is an apparent contradiction to conventional understanding of solidification behavior because the interior of the sample contains less crystalline material than the exterior. However the cooling rate is sufficently slow in during the casting process to allow for bulk diffusion to occur during cooling. This effect causes the exterior of the sample to become partially crystalline and causes the composition of the remaining liquid becomes more Nd-rich as is required by mass balance. It appears that the glass-forming ability of the remaining liquid composition is greater than that of the initial composition.

The origin of this behavior is related to the equilibrium ternary phase diagram and will be discussed in another paper.

In summary, we have shown that the use of x-ray diffraction analyses can reveal clearly the presence of microsized crystallites in the outermost and innermost regions of the suction-cast rods of $Nd_{60}Fe_{30}Al_{10}$ BMG-type alloy. The effects of solidification-related changes in the liquid composition are demonstrated to significantly alter the glass-forming ability of the melt. The attendant magnetic measurements demonstrate that the cast samples are inhomogenous not only along their thickness but also along their height, *i.e.*, along the distance from the bottom of the mold to the top. The differences in the degrees of crystallinity and phase proportions found in the samples of identical composition but processed by the two different solidification methods — melt-spinning and suction casting — lie in the time scale over which diffusion is possible during solidification.

ACKNOWLEDGMENTS

Research was performed under the auspices of the U.S. Dept. of Energy, at Ames Laboratory under Contract No. W-7405-ENG-82 and at Brookhaven National Laboratory under contract No. DE-AC02-98CH10886. Support from the East Asia and Pacific Program of the Division of International Programs, N.S.F., is gratefully acknowledged. The IMS authors are in debt to Dr. P.V. Phuc and Dr. J. Ding for the XRD measurements as well as for valuable discussions. They also appreciate the financial support from a NCST grant and from SAREC/Sida Project on Rare Earth Materials.

REFERENCES

[1]. A. Inoue, T. Zhang and A. Takeuchi, Mat. Trans. JIM **37** (2) (1996) 99.

[2]. A. Inoue, T. Zhang and A. Takeuchi, IEEE Trans. Magn. **33** (1997) 3814;

[3]. K. V. Rao, Dept. of Condensed Matter Physics, The Royal Institute of Technology, Stockholm, Sweden, presented at the Spring MRS Research Society Meeting, San Francisco, CA April 5-8, 1999.

[4]. A. Inoue, T. Zhang and A. Takeuchi, Met. and Mat. Trans. **29A** (1998) 1779.

[5]. Y. Li, J. Ding, S. C. Ng and X . Z. Wang, J. Magn. Magn. Mat. **187** (1998) 273.

[6]. J. Ding, Y. Li and X. Z. Wang, J. Phys. D: Appl. Phys. **32** (1999) 713.

[7]. A. S. O'Connor, L. H. Lewis, R. W. McCallum, K. W. Dennis, M. J. Kramer, D. T. K. Anh, N. H. Dan, N. H. Luong and N. X. Phuc, Proc. of the 16th Intern. Workshop on Rare Earth Magnets, Sendai, Japan, Sept. 10-14 (2000) 475.